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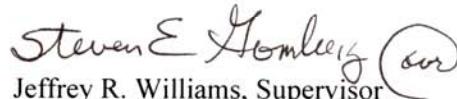
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YUCCA MOUNTAIN - SUPPLEMENTAL RESPONSES TO REQUEST FOR ADDITIONAL INFORMATION - SAFETY EVALUATION REPORT VOLUME 3 - POSTCLOSURE CHAPTER 2.2.1.3.4 - RADIONUCLIDE RELEASE RATES AND SOLUBILITY LIMITS, SET 2 (DEPARTMENT OF ENERGY'S SAFETY ANALYSIS REPORT SECTION 2.3.7)

- Reference 1: Ltr, Sulima to Williams, dtd 4/7/2009, "Yucca Mountain – Request for Additional Information – Safety Evaluation Report, Volume 3 – Postclosure Chapter 2.2.1.3.4 –Radionuclide Release Rates and Solubility Limits, Set 2 –(Department of Energy's Safety Analysis Report Section 2.3.7)"
- Reference 2: Ltr, Williams to Sulima, dtd 5/12/09, "Yucca Mountain – Request for Additional Information – Safety Evaluation Report, Volume 3 – Postclosure Chapter 2.2.1.3.4 –Radionuclide Release Rates and Solubility Limits, Set 2 –(Department of Energy's Safety Analysis Report Section 2.3.7)"

The purpose of this letter is to submit the U.S. Department of Energy's (DOE) supplemental response to Request for Additional Information (RAI) Number 9 identified in reference 1 above. The original response to that RAI was provided by reference 2 above. Subsequently, it was discovered that the units for bulk residence time were incorrectly presented in Equation 5, where time in years was shown in the denominator. The corrected equation places units of time in the numerator of that equation. This error did not affect any of the response discussion or conclusions. The entire response is provided for your convenience as an enclosure to this letter.

There are no commitments in the enclosed RAI response. If you have any questions regarding this letter, please contact me at (202) 586-9620, or by email to jeff.williams@rw.doe.gov.


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Regulatory Affairs Division
Office of Technical Management

OTM:CJM-0233

Enclosure:

Corrected response to RAI Volume 3, Chapter 2.2.1.3.4, Set 2, Number 9



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RAI Volume 3, Chapter 2.2.1.3.4, Second Set, Number 9:

Demonstrate that the procedure for establishing the range of forward rate constants from sorption data does not result in EBS releases of plutonium being underestimated.

Basis: The range of forward rate constants for sorption of americium and plutonium onto corrosion products was estimated from plutonium sorption experiments in SNL (2007a). The rate constants were calculated from the plutonium sorbed fraction at the end of the experiments. Rapid early uptake of plutonium was ignored when estimating the rates. Had the rates been estimated from the early part of the time series, the estimated rate constants would be larger. Larger forward rate constants would result in larger transfer of radionuclide mass to mobile corrosion product colloids and possibly to larger colloid-associated plutonium and americium releases from the EBS. In addition, because rate constants for desorption from stationary corrosion products are proportional to the forward rate constants in the abstraction, underestimating the forward rate constants would also result in the desorption rates being underestimated, with potential implications for EBS releases of plutonium and americium. This information is needed to verify compliance with 10 CFR 63.21(c)(11) and (12), and 63.114(b).

1. RESPONSE

This response demonstrates that the procedure for establishing the range of forward rate constants from sorption data does not result in Engineered Barrier System (EBS) releases of plutonium being underestimated and that the method used was appropriate for the EBS environment. This RAI response should be read in conjunction with the response to RAI 3.2.2.1.3.4-2-008, because both responses discuss the forward rate constant for sorption of plutonium and americium onto iron oxyhydroxide surfaces and provide complementary justifications for the appropriateness of the range used in the performance assessment.

1.1 USE OF THE FORWARD RATE CONSTANT IN THE TSPA MODEL

As described in Section 6.5.1.2 of *EBS Radionuclide Transport Abstraction* (SNL 2007a, Equations 6.5.1.2-14, -15, and -20), the forward rate constant is used to calculate (1) the plutonium and americium sorption/desorption rate onto stationary corrosion products (CP) and (2) the rate of sorption onto iron oxyhydroxide colloids in the EBS.

The response to RAI 3.2.2.1.3.4-2-001 details under what EBS environmental conditions the forward rate constant is based on experimental data, and under what conditions it is based on the target flux out ratio.¹ The response below deals specifically with the conditions and realizations

¹ The target flux out is the ratio of radionuclide mass associated with colloids (reversible and irreversible) to radionuclide mass associated with colloids and dissolved radionuclide mass (dimensionless). It is a sampled uncertain variable, CFLUXOUT (SAR Table 2.4-11).

for which the log-uniform experimental range of 0.002 to 0.05 m³/(m² yr) is applicable (SNL 2007a, Section 6.3.12.2).

1.2 JUSTIFICATION OF THE FORWARD RATE CONSTANT RANGE

The justification for the use of the late-time sorbed fraction in the Lu et al. (2000) experiments (SNL 2007b) is that the EBS environmental conditions and corresponding residence times are more in line with the late time experimental conditions. The very early time sorption rates on the time scale of minutes, were not ignored, but, rather were considered inappropriate for directly representing radionuclide transport in the EBS environment. Furthermore, the sorption represented by the very early time rate is captured in the time-averaged rates used in the model (SNL 2007b, Section 6.3.12.2).

When Equations 6.5.1.2-3, -7, -8, -14, -15, and -32 in *EBS Radionuclide Transport Abstraction* (SNL 2007a) are nondimensionalized in time and in one spatial dimension, two dimensionless groups, the forward and backward rate Damköhler numbers, N_{Da}^f and N_{Da}^r , will appear in those equations:

$$N_{Da}^f = \rho_b \bar{s}_{CP} k_{if} \tau \quad (\text{Eq. 1})$$

$$N_{Da}^r = \rho_b \bar{s}_{CP} k_{ir} \tau \quad (\text{Eq. 2})$$

where ρ_b is the dry bulk density of the stationary solid matrix (kg/m³ bulk volume), \bar{s}_{CP} is the specific surface area of iron oxyhydroxide corrosion products (CP) [m² CP/kg CP], k_{if} is the kinetic forward rate constant for species i (m³ water/(m² CP yr)), k_{ir} is the kinetic backward rate constant for species i (m³ water/(m² CP yr)), and τ is the residence time per bulk volume (yr or m³ bulk volume/(m³ water volume yr)), defined as:

$$\tau = \frac{V_b}{Q_{WP}} \quad (\text{Eq. 3})$$

where V_b is the bulk volume in the corrosion products cell (m³ bulk volume) and Q_{WP} is the volumetric fluid flow rate through the waste package (m³ water/yr).

The Damköhler number represents the ratio of the rate of reaction to the rate of transport and, if both N_{Da}^f and N_{Da}^r are greater than about 10, local equilibrium conditions are present (Sevougian et al. 1993) and greater values of the rate constant would not change the transport behavior.

An example of the approximate magnitude of the forward rate Damköhler number can be given for the igneous intrusion modeling case simulation for failed commercial spent nuclear fuel waste packages in a seeping environment in Percolation Subregion 3. The igneous intrusion modeling case is considered because it is one of the two most risk-significant cases and it is the modeling case for which colloid transport has the greatest potential to be important (due to stable

colloid suspensions and advective transport conditions). The dry bulk density of the stationary corrosion products, ρ_b (i.e., the mass of corrosion products per bulk volume), is the product of the solid-phase volume fraction (or $1 - \phi$, where ϕ is the porosity in m³ void/m³ bulk volume) and the matrix density of the corrosion products, ρ_{ma} (kg/m³ CP). The porosity is 0.4 (SNL 2007a, Section 6.5.2.1.1.2) and the corrosion products matrix density is a linear average of the fraction of each component in the corrosion products domain multiplied by its matrix density. The components and their matrix densities are goethite (4,260 kg/m³), hydrous ferric oxide (HFO) (3,960 kg/m³), Cr₂O₃ (5,220 kg/m³), and NiO (6,720 kg/m³) (SNL 2007a, Table 6.5-6). The fraction of each component in the corrosion product domain is determined using the estimated masses of each of the steel components from Table 6.3-8 (SNL 2007a), 25,600 kg for iron oxides (FeOOH), 6,100 kg for Cr₂O₃, and 3,750 kg for NiO to the total corrosion products mass, 35,400 kg. Given the mean sampled proportions of 0.625 for goethite in iron oxides (SNL 2007a, Table 6.5-7, Item 5) and 0.375 for HFO (1 – goethite abundance), this gives an average matrix density of 4,610 kg/m³. Thus,

$$\rho_b = \rho_{ma}(1 - \phi) = 4610 \times 0.6 = 2770 \text{ kg CP/m}^3 \text{ bulk volume} \quad (\text{Eq. 4})$$

The mean specific surface area of iron oxyhydroxide corrosion product, \bar{s}_{CP} , is 135.5 m² CP/g and the mean forward rate constant, k_{if} , is equal to 0.015 m³ water/(m² CP yr) (SNL 2007a, Section 6.3.12.2). The product of these three terms, $\rho_b \bar{s}_{CP} k_{if}$, equals $5.63 \times 10^6 \text{ m}^3 \text{ water}/(\text{m}^3 \text{ bulk volume yr})$. The bulk residence time at 1,000,000 years (see Equation 3) can be computed from the ratio of the average water volume in the corrosion products, V_w , and the average volumetric water flux through the waste package, Q_{WP} (both of which are outputs from the TSPA model):

$$\tau = \frac{V_b}{Q_{WP}} = \frac{V_w}{\phi S_w Q_{WP}} = \frac{5.27 \text{ m}^3}{(0.4)(1.0)(0.945 \text{ m}^3/\text{yr})} = 13.94 \text{ m}^3 \text{ bulk volume}/(\text{m}^3 \text{ water/yr}) \quad (\text{Eq. 5})$$

where S_w is the water saturation and is equal to 1 for the igneous intrusion case. (Note that the pore residence time, i.e., the residence time per unit pore volume, is simply $V_w/(S_w Q_{WP})$.) Multiplying the above value of bulk residence time, τ , by the value of $\rho_b \bar{s}_{CP} k_{if}$ results in a value of the dimensionless forward rate Damköhler number, N_{Da}^f , equal to 7.85×10^7 at the time of 1,000,000 years.

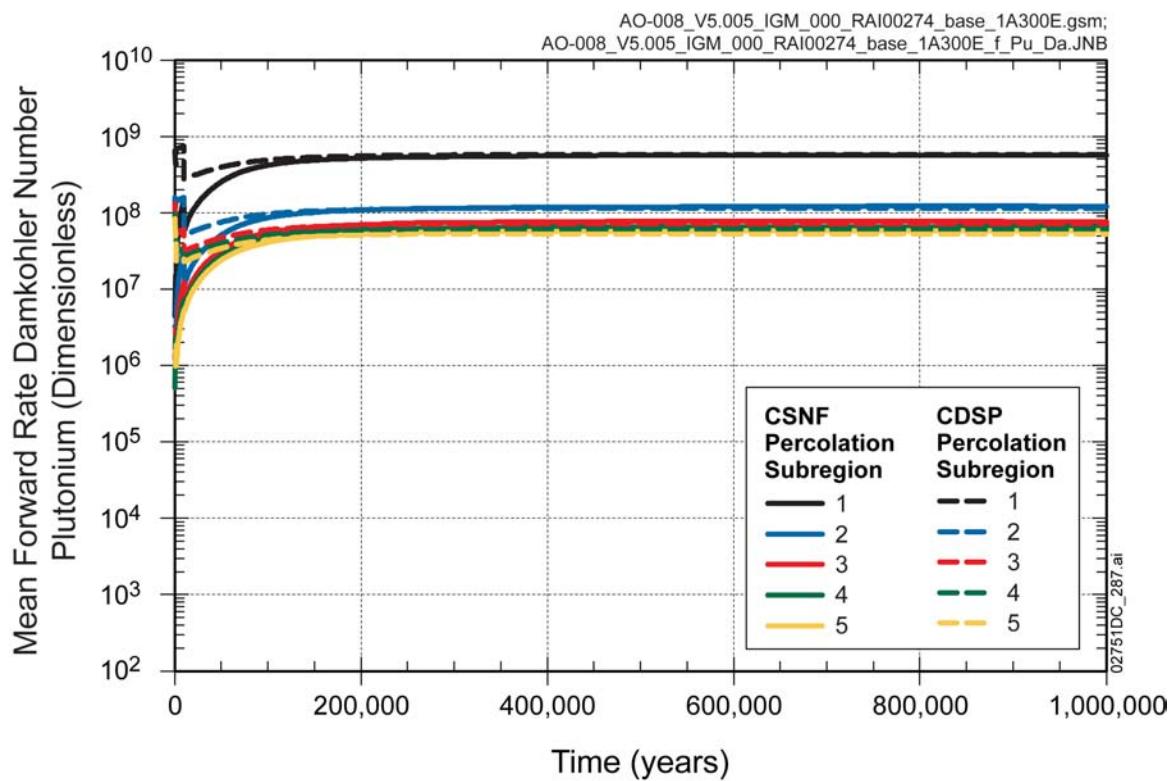
Figures 1 and 2 show the range and time histories of the mean Damköhler numbers over the various percolation subregions for the igneous intrusion modeling case. The backward rate Damköhler number, N_{Da}^r , is equal to the forward rate Damköhler number divided by the K_d computed from the surface complexation model for the corresponding radionuclide (or, more specifically, by $K_d \rho_b / \phi S_w$ in order to get the correct units on k_{ir}). The forward and backward rate Damköhler numbers for plutonium are greater than 10, so the use of a higher value for the rate constant (even if it were appropriate) would not change the transport behavior with respect

to sorption onto the stationary corrosion products. Thus, EBS releases are not underestimated with respect to this process.

With regard to irreversible sorption onto iron oxyhydroxide colloids, there is a long contact time of colloids with solutes in aqueous-phase fluid packets, because the colloids travel with the solution. Thus, a rate constant from the early part of the experimental time series is inappropriate for colloids, and the rate constant computed from the end of the experiments is appropriate. This is discussed in greater detail in the response to RAI 3.2.2.1.3.4-2-008.

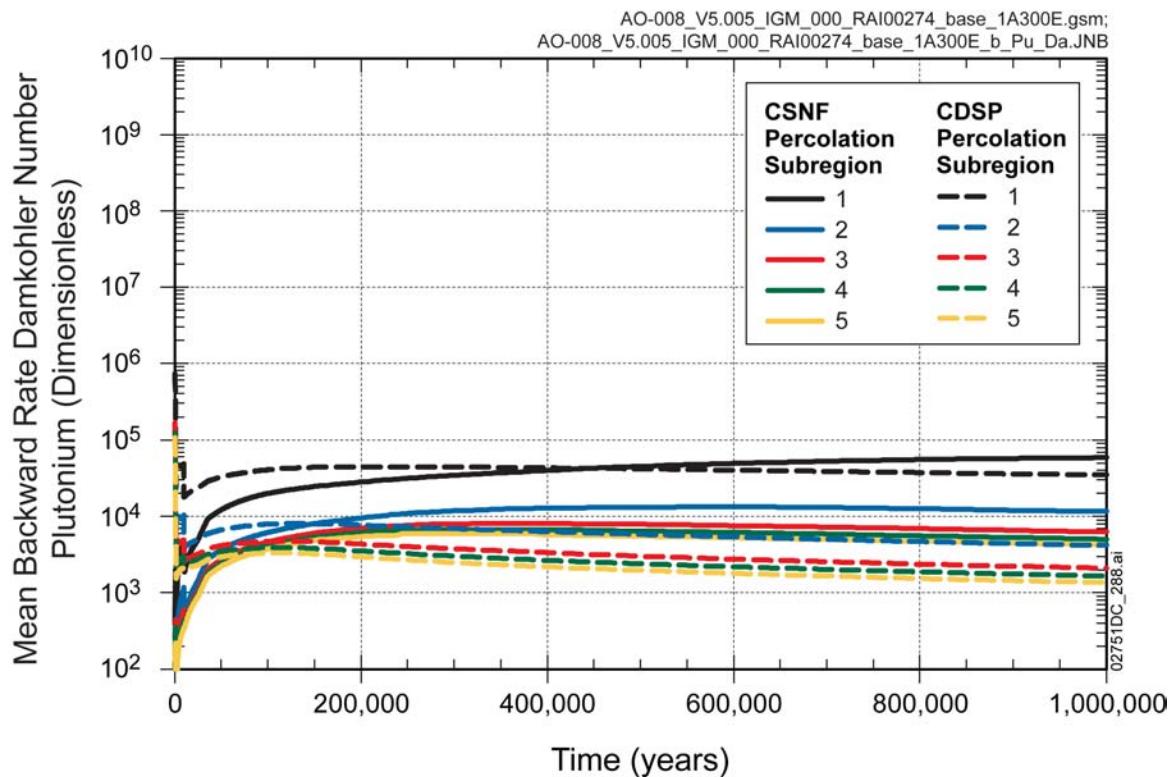
1.3 SUMMARY

The late time sorbed fractions from the plutonium sorption experiments are appropriate for establishing a range of forward rate constants that are applicable under repository environmental conditions, and therefore the sampled range does not underestimate plutonium releases from the EBS. Based on residence times and Damköhler numbers in the EBS, the long time rate constants from the experimental data are appropriate and, even if higher rate constants were used, transport behavior would not be affected because local equilibrium conditions apply even at the lower values of the rate constant.



NOTE: CDSP = codisposal; CSNF = commercial spent nuclear fuel.

Figure 1. Mean Forward Rate Damköhler Numbers for Plutonium from the Igneous Intrusion Modeling Case with an Event Time at 250 Years, in the Five Percolation Subregions for Commercial SNF and Codisposal Waste Packages



NOTE: CDSP = codisposal; CSNF = commercial spent nuclear fuel.

Figure 2. Mean Backward Rate Damköhler Numbers for Plutonium from the Igneous Intrusion Modeling Case with an Event Time at 250 Years, in the Five Percolation Subregions for Commercial SNF and Codisposal Waste Packages

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

Lu, N.; Conca, J.; Parker, G.R.; Leonard, P.A.; Moore, B.; Strietelmeier, B.; and Triay, I.R. 2000. *Adsorption of Actinides onto Colloids as a Function of Time, Temperature, Ionic Strength, and Colloid Concentration, Waste Form Colloids Report for Yucca Mountain Program (Colloid Data Summary from 1999 to 2000 Research)*. LA-UR-00-5121. Los Alamos, New Mexico: Los Alamos National Laboratory. ACC: MOL.20031204.0108.

Sevougian, S. D.; Schechter, R. S.; and Lake, L.W. 1993. "Effect of Partial Local Equilibrium on the Propagation of Precipitation/Dissolution Waves." *Industrial & Engineering Chemistry Research*, 32, (10), 2281-2304. Washington, D.C.: American Chemical Society.

SNL (Sandia National Laboratories) 2007a. *EBS Radionuclide Transport Abstraction*. ANL-WIS-PA-000001 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20071004.0001.

SNL 2007b. *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary*. MDL-EBS-PA-000004 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20071018.0019.^a

NOTE: ^aProvided as an enclosure to letter from Williams to Sulima dtd 02/17/2009. "Yucca Mountain – Request for Additional Information Re: License Application (Safety Analysis Report Section 2.1), Safety Evaluation Report Volume 3 – Postclosure Chapters 2.2.1.1 and 2.2.1.3.7 – Submittal of Department of Energy Reference Citations."